Submitted for publication.

Determination of the ^{85}Rb ng-series quantum defect by resonant energy transfer spectroscopy of cold Rydberg atoms

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(Dated: August 22, 2006)

Resonant energy transfer between cold Rydberg atoms was used to determine Rydberg atom energy levels, at precisions approaching those obtainable in microwave spectroscopy. Laser cooled ^{85}Rb atoms from a magneto-optical trap were optically excited to $32d_{5/2}$ Rydberg states. The two-atom process $32d_{5/2} + 32d_{5/2} \rightarrow 34p_{3/2} + 30g$ is resonant at an electric field of approximately 0.3 V/cm. This process is driven by the electric dipole-dipole interaction, which is allowed due to the partial f character that the g state acquires in an electric field. The experimentally observed resonant field, together with the Stark map calculation is used to make a determination of the ^{85}Rb ng-series quantum defect: $\delta_g(n = 30) = 0.00405(6)$.

PACS numbers: 32.80.Rm, 34.20.Cf, 39.30.+w, 32.30.-r

I. INTRODUCTION

Energy can be transferred more rapidly between two atoms when the process is *resonant*: one atom gives its energy to another, without a change in the kinetic energy of either atom. Although resonant energy transfer often relies on serendipity, the large Stark shifts of Rydberg atoms may allow the resonant condition to be exactly obtained by application of an electric field [1]. The resonant energy transfer process between Rydberg atoms is driven by the electric dipole-dipole interaction. Transition dipole moments between nearby Rydberg states can be large, typically scaling as n^2, where n is the principal quantum number [2]. As a consequence, resonant energy transfer collision cross-sections scale as n^4, and are large compared to typical collision cross-sections for less excited atoms – usually by many orders of magnitude [1, 2].

Stoneman *et al.* [3] pointed out that the fields at which resonant energy transfer occurs may – under certain conditions – be used for precise Rydberg energy level spectroscopy. They examined the resonant energy transfer process: $29s + 29d \rightarrow 29p + 28p$, in potassium. Since the shifts of the Rydberg energy levels with field are relatively well-understood, they were able to use the resonance fields to determine improved zero field atomic energy levels. In particular, they determined a substantially more precise value for the quantum defect of the np-series of potassium.

The range of electric fields or "width" over which significant resonant energy transfer can be observed is important in this type of experiment, as it dictates the precision to which line centers can be determined. By reducing the spread in the velocity distribution of their sample Stoneman *et al.* [3] were able to significantly narrow the width of their observed resonances.

With the development of laser cooling, Rydberg atom samples with much narrower velocity distributions could be studied [4, 5]. In general, the resulting narrowness of the resonant energy transfer resonances has not been exploited for spectroscopic purposes. Instead, work has concentrated on the dynamical aspects of resonant energy transfer between cold Rydberg atoms. For example, Carroll *et al.* [6] have recently studied how the relative orientation of the electric field with respect to the spatial orientation of two Rydberg atoms influences their dipole-dipole interaction. They used cold Rb atoms obtained from a magneto-optical trap (MOT) to study the process: $32d + 32d \rightarrow 34p + 30k$, where $30k$ represents the Stark-states centered around $n = 30$. The Stark states are superpositions of high angular momentum states ($\ell \gtrsim 4$ in this case) exhibiting energy shifts which are linear in the applied electric field strength. Strong resonant energy transfer was observed between 1 to 6 V/cm, and was fully explained by a Stark map calculation, which took into account the energy shifts of the participating atoms. However, a series of peaks at approximately 0.5 V/cm were not reproduced in the calculations (these peaks were not relevant to the main point of their paper).

In this paper we demonstrate that the resonances observed at low field are due to the process:

$$32d + 32d \rightarrow 34p_{3/2} + 30g \qquad (1)$$

which is an allowed electric dipole-dipole interaction, due to the f character that the g state acquires in the electric field. In the spirit of the work of Stoneman *et al.* [3] we use the observed resonant fields together with a Stark map calculation to determine the ng-series quantum defect.

Han *et al.* [7] have reported a preliminary observation of two-photon $(n + 2)d - ng$ microwave transitions in ^{85}Rb. However, we are unaware of any other work on the ng Rydberg series of Rb.

II. EXPERIMENT

The essence of the experiment is to measure the electric fields required for efficient resonant energy transfer

between cold $32d_{5/2}$ ^{85}Rb atoms. Some details of our apparatus have been previously described [8]. A standard vapor cell MOT is used as the source of cold ^{85}Rb atoms. These atoms are excited to $32d_{5/2}$ Rydberg states using a two-photon, two-color process with 480 nm light, and the nearly resonant, red-detuned 780 nm light used for cooling and trapping. The 480 nm light is obtained by frequency doubling a 960 nm cw ring Ti:sapphire laser, which is frequency stabilized using the technique described in Ref. [9]. The experiment is done at a 10 Hz repetition rate, with an acousto-optic modulator used to produce pulses of light.

For energy level determination from resonant energy transfer spectroscopy it is critical to zero out any stray electric fields, and then be able to apply accurately known electric fields. Two stainless steel electrode plates, separated by 36 mm, are located to either side of the trapped atoms. By varying the voltages on these plates and the voltage of the Rb dispenser source with respect to the grounded chamber, it is possible to compensate the stray electric field in all three directions. The single-photon transition $48s_{1/2} - 48p_{1/2}$ in ^{85}Rb is used for this purpose. This general procedure has been described by Osterwalder and Merkt [10]. Since the single photon transition is sensitive to magnetic fields, the inhomogeneous magnetic field necessary for MOT operation is shut-off prior to photoexcitation to Rydberg states. At the time of the experiment the residual magnetic field inhomogeneity over the sample is less than 17 mG. The details of this shutting off procedure are presented in Ref. [8].

Once the stray electric field is compensated, the plate voltages may be used to produce deliberate fields. We calibrate these fields by observing the shifted frequencies of the $48s_{1/2} - 48p_{1/2}$ transition and comparing them to the frequencies expected from a Stark map calculation [11]. The Stark map calculation uses the most recent quantum defect data for ^{85}Rb [7, 12]. Although quantum defects for $\ell > 3$ are not known, these do not significantly influence the calculated transition frequencies.

To observe resonant energy transfer between Rydberg atoms with known electric fields, we produce the fields in exactly the same way as during calibration using the $48s_{1/2} - 48p_{1/2}$ transition. The inhomogeneous magnetic field is also shut-off in an identical manner. After a light excitation pulse of 1 μs duration, the electric field is ramped from zero to the desired value in 160 ns. The atoms may interact during a waiting period of 21 μs.

Final Rydberg state populations after the waiting period are analyzed by selective field ionization (SFI) [2]. In this case SFI is only able to distinguish states differing by energies equivalent to a change of approximately one principal quantum number. The $34p$ Rydberg states are ionized at a lower field than both the $32d_{5/2}$ initial states and $30g, k$ final states. By changing the applied electric field and recording the $34p$ population, resonant energy transfer spectra may be obtained (see Fig. 1c). For comparison we have also recorded analogous spectra at one lower n (Fig. 1a) and one higher n (Fig. 1e).

III. ANALYSIS

The striking difference in the three spectra presented in Fig. 1 is the presence of the low-field resonances in the $32d_{5/2}$ case, but not in the others. Carroll *et al.* have also observed similar resonances in $32d$ (Ref. [6]), but not for $31d$ (Ref. [13]). Energetics [7, 12] rule out the process: $32d_{5/2} + 32d_{5/2} \rightarrow 34p_{3/2} + 30f$ as a possible explanation for these low-field resonances. The final state $34p_{3/2}$+$30f$ is at an energy less than the initial state in zero electric field, and this difference increases with increasing field, due to the f state polarizability.

There are no processes involving the well-characterized low-angular momentum Rydberg states ($\ell \leq 3$) of Rb which would account for the low field resonances in Fig. 1c. We need to consider higher angular momentum states. The non-penetrating high angular momentum states of Rydberg atoms have quantum defects which scale like $1/\ell^5$ (see, for example, Ref. [2]). Thus a rough estimate of the ng-series quantum defect may be obtained from the nf-series quantum defect [7]: $\delta_g \approx \delta_f (3/4)^5 \approx 0.004$. A preliminary calculation with this estimated quantum defect shows that a resonance will occur for the process: $32d_{5/2}(m_j = 1/2) + 32d_{5/2}(m_j = 1/2) \rightarrow 34p_{3/2}(m_j = 1/2) + 30g_{7/2}(m_j = 1/2)$ at an electric field of 0.31 V/cm. This is very close to the lowest field resonance observed in Fig. 1c. However for the $31d_{5/2}$ case, the energy of the two-atom state $33p_{3/2} + 29g$ is less than the initial state $31d_{5/2} + 31d_{5/2}$, and this difference increases with increasing field (see Fig. 1b). This is consistent with the lack of observed low-field resonances in the $31d_{5/2}$ spectrum (Fig. 1a).

In the case of the $33d_{5/2}$, the electric field does shift $35p_{3/2} + 31g$ closer in energy to $33d_{5/2} + 33d_{5/2}$. However, the $31g$ state reaches the $n = 31$ Stark manifold at approximately the same field (Fig. 1f). This is consistent with the experimental observations in Fig. 1e.

The calculations in Fig. 1 consider only $m_j = 1/2$ magnetic sublevels. However, electric fields break the magnetic sublevel energy degeneracy; therefore, there should be different resonance fields corresponding to the different possible magnetic sublevels for the states in Eq. 1. The largest splittings are due to the magnetic sublevels of $30g$. The four resonances observed at low fields in Fig. 1c correspond to $m_j = 1/2, 3/2, 5/2,$ and $7/2$. The splittings due to the magnetic sublevels of the other states are much less than the widths of each of these four peaks.

All four observable resonances should be explainable by the same ng-series quantum defect (δ_g). Figure 2a illustrates calculated resonance fields as a function of a variable δ_g, centered around the estimated $\delta_g \approx 0.004$ (see above). As illustrated in Fig. 2, all four resonances are consistent with the same quantum defect, to better than the widths of the resonances. We consider this as strong evidence that the process in Eq. 1 is the explana-

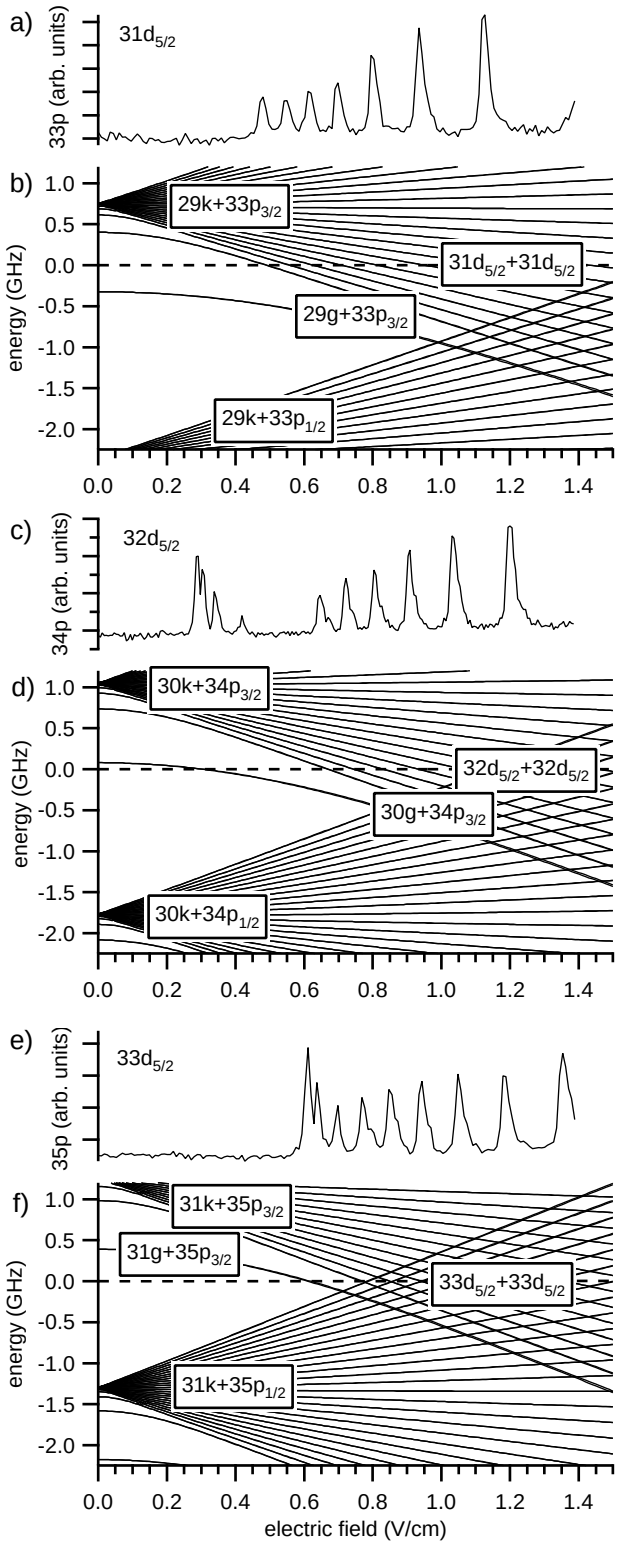

FIG. 1: (a,c,e) Resonant energy transfer spectra with atoms initially in the $31d_{5/2}$, $32d_{5/2}$ and $33d_{5/2}$ states. (b,d,f) Calculated total energy of several two atom states, relative to the zero field energy of $nd_{5/2} + nd_{5/2}$ where $n = 31, 32, 33$ for (b), (d) and (f) respectively. The calculations follow the procedures of Zimmerman *et al.* [11], using $\delta_g = 0.004$ and for $\ell > 4$, $\delta_\ell = \delta_g(4/\ell)^5$.

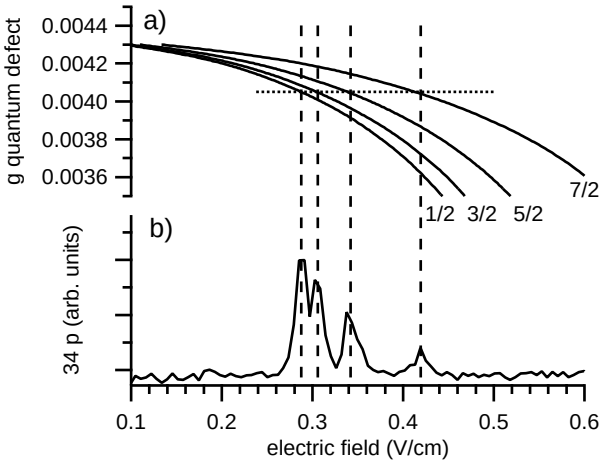

FIG. 2: a) Resonance fields calculated for variable δ_g (vertical axis). The four lines correspond to different magnetic sub-levels (m_j) for the $30g$ state. The calculated fields are shown on the horizontal axis to allow a direct comparison to the experimental data below. b) The same $32d_{5/2} + 32d_{5/2}$ resonant energy transfer spectrum as Fig. 1c), shown on an expanded scale.

tion for the low field resonances.

As Fig. 2 suggests, we can extract an estimate of the quantum defect from each one of these four resonances. The uncertainty may also be estimated for each of these experimentally determined quantum defects from the derivatives $\partial\delta_g/\partial F$ evaluated at the estimated quantum defects (where F is the electric field strength). The electric field zeroing and calibration procedure is estimated to have a 1σ uncertainty of less than 30 mV/cm at the resonance fields. From the four resonances we determine $\delta_g(n = 30) = 0.00405(6)$, which is consistent with a preliminary report of $\delta_g = 0.00400(9)$ from Han *et al.* [7].

As demonstrated, energetics strongly support Eq. 1 as the explanation for the low-field resonances. However, what drives this process? Resonant energy transfer between Rydberg atoms is typically a dipole-dipole interaction, with the coupling between the two-atom states given by:

$$\hat{V}_{dd} = \frac{\vec{\mu}_{\rm A} \cdot \vec{\mu}_{\rm B} - 3(\vec{\mu}_{\rm A} \cdot \vec{n})(\vec{\mu}_{\rm B} \cdot \vec{n})}{R^3_{\rm AB}} \qquad (2)$$

where $R_{\rm AB}$ is the separation of the two atoms, $\vec{n}$ is a unit vector pointing between them, and $\vec{\mu}_{\rm A}$ and $\vec{\mu}_{\rm B}$ are *transition* dipole moments evaluated on each of the individual atoms (A and B). In zero electric field the process given in Eq. 1 cannot be driven by the dipole-dipole interaction, since $\langle 32d_{5/2}|\vec{\mu}|30g_{7/2}\rangle = 0$, given the $\Delta\ell = \pm 1$ selection rule for transition dipole moments. However, when a weak electric field is applied, the ℓ quantum number is no longer good. Instead, it serves to label the dominant character of a state until it reaches the Stark manfold. In particular, the mixing of $30g$ with $30f$ gives $\langle 32d_{5/2}m_j{=}1/2|\mu_z|30g_{7/2}m_j{=}1/2\rangle = 17\ q_e a_0$

at 0.3 V/cm, where $q_e a_0$ is the atomic unit of electric dipole moment. The $32d_{5/2}$ to $34p_{3/2}$ coupling is allowed in zero field, and is much stronger. For example at 0.3 V/cm, $\langle 32d_{5/2} m_j{=}1/2|\mu_z|34p_{3/2} m_j{=}1/2\rangle = 357\, q_e a_0$ (which differs little from its zero field value).

It is interesting to compare the expected signal strengths for the resonance in Eq. 1 with other resonances that have been studied in similar experimental set-ups. For example, if we compare our transition dipole moments to those for the ^{85}Rb, $33s_{1/2} + 25s_{1/2} \rightarrow 24p_{1/2} + 34p_{3/2}$ process observed in Ref. [4], we would expect to have to wait approximately 10 times longer to see the equivalent amount of collision signal (at the same density). Since typical waiting times are 3 μs in Ref. [4] *vs.* 21 μs in our case, this is consistent.

In the quasi-static picture [4] the collision signal is expected to scale with the square of the density for a dipole-dipole interaction, and to a higher power for higher-order multipole processes (for small collision signals). Experimentally, we observe square law behavior. It does not appear necessary to consider higher-order multipole interactions (see, for example, Ref. [2]).

IV. CONCLUDING REMARKS

Resonant energy transfer spectroscopy has been demonstrated as a useful tool for the determination of unknown energy levels differences. In particular, prior to the work of Han *et al.* [7], no spectroscopy of the Rb ng-series had been performed.

Although resonant energy transfer spectroscopy is not a general technique for the determination of energy levels, there may be more possibilities for the observation of resonances than previously thought. In particular, at first glance the resonance studied in this paper (Eq. 1) does not appear to be allowed by the dipole-dipole interaction. However, the mixing of the g-state with the nearby f-state allows this process to be observed. It is expected that similar scenarios exist in Rb and other atoms.

Microwave transitions have been used to calibrate the applied electric fields in this work. However, there is a great deal of flexiblity in the particular transition (and corresponding microwave frequency) that can be used for this calibration. A precise microwave source is not required for the actual spectroscopic measurement. This may be considered to be a significant advantage.

We thank the authors of Ref. [7] for providing us with a preprint prior to publication and acknowledge useful conversations with T. F. Gallagher, M. Noel and J. D. Carter. We thank D. Vagale for assistance with the Stark map calculations. This work was supported by NSERC, CFI, and OIT.

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